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(21) International Application Number: PCT/GB91/00930 (22) International Filing Date: 11 June 1991 (11.06.91) (30) Priority data: 9013307.5 14 June 1990 (14.06.90) GB (71) Applicant (for all designated States except US): ALLIED COLLOIDS LIMITED [GB/GB]; P.O. Box 38, Low Moor, Bradford, West Yorkshire BD12 0JZ (GB). (72) Inventors; and (75) Inventors/Applicants (for US only) : FARRAR, David [GB/GB]; 2 Pennyhill Drive, Clayton, Bradford, West Yorkshire BD14 6NH (GB). HAWE, Malcolm [GB/GB]; 7 Portland Close, Lindley, Huddersfield, West Yorkshire HD3 4BS (GB). DOUGAL, Joyce, Collin [GB/GB]; 51 Willow Rise, Thorpe Willowghby, Selby, North Yorkshire YO8 9PP (GB).			(74) Agent: LAWRENCE, Peter, Robin, Broughton; Gill Jennings & Every, 53 to 64 Chancery Lane, London WC2A 1HN (GB). (81) Designated States: AT (European patent), BE (European patent), CA, CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (European patent), GB (European patent), GR (European patent), IT (European patent), LU (European patent), NL (European patent), SE (European patent), US. Published With international search report.
(54) Title: POLYMERIC FILMS FOR FORMING CAPSULE SHELLS			
(57) Abstract <p>A composition suitable for forming capsule shells is a water soluble synthetic film-forming polymeric composition of cationic and anionic moieties, and is preferably a blend of cationic polymer with anionic polymer. The capsule shells formed of such a composition is suitable for containing a range of non-aqueous fillings ranging from bath additives such as bath oil to hazardous chemicals such as pesticides.</p>			

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POLYMERIC FILMS FOR FORMING CAPSULE SHELLS

This invention relates to polymeric films which are suitable for forming shells or capsules for containing various non-aqueous liquids or solids.

5 Depending on the various uses of the capsule shells, the required properties may vary. At present, capsules are usually made from gelatin. However there are various reasons why it is sometimes undesirable to use a naturally occurring material such as gelatin, for instance because of
10 its variable quality or because of a public preference to use a synthetic material rather than an animal-based material. It is known to form capsules from single homopolymeric materials such as polyvinyl alcohol. However, it is difficult with these polymeric compositions
15 to produce a product having all the desired properties for each alternative use.

The walls of gel capsules must have high mechanical strength and stability, and resistance to the substance which will be contained in the capsule shell. It is often
20 necessary that the capsule shells should dissolve rapidly in water. However, the solubility properties can vary considerably from one requirement to another, for example the temperature of the aqueous solution in which the shell must dissolve may vary considerably. For example if
25 capsules are filled with pesticides or herbicides the capsule walls must dissolve in cold water, so that the pesticide may be readily diluted for application to the land, whereas when the contained substance is bath oil, rapid solubility must be achieved for warm water. In
30 addition, for example, if the capsule shells are used for containing pharmaceutical preparations or cosmetic products such as bath oils, the gel capsule must be aesthetically acceptable and therefore it is beneficial if the film-forming polymeric material can be dyed and/or can be
35 mixed with pearling agents.

The known homopolymeric systems for making capsules have very low controllability and it is difficult to vary

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th properties to meet th different requirements of capsule shells for various uses. In particular, no synthetic film forming system has previously been proposed that provides a satisfactory combination of flexibility and non-stickiness in the presence of ambient humidity, mechanical strength, and total rapid dissolution in water.

This invention relates to a film-forming polymeric system suitable for preparing capsule shells which can be varied easily according to the particular requirements of the filling material and , especially, to a system that is flexible, non-sticky and completely water soluble.

According to the present invention there is provided a polymeric composition suitable for forming a capsule shell for containing a substantially non-aqueous substance characterised in that the composition comprises a water soluble synthetic film-forming polymeric composition of cationic and anionic moieties.

The invention also provides a capsule shell in which at least part of the shell is formed from such a composition. For instance the total capsule may be formed from two shells and at least one, and preferably both, of the shells are formed of the composition. A shell may be formed of two parts, one part being formed of the defined composition and the other part being formed from some other capsule-forming material. The invention also provides filled capsules in which at least part of the capsule shell, and preferably all the capsule shell, is formed from the composition.

The synthetic film forming polymeric composition can comprise an amphoteric polymer formed by co-polymerisation of anionic ethylenically unsaturated monomer with cationic ethylenically unsaturated monomer and, optionally, nonionic ethylenically unsaturated monomer.

Preferably the cationic and anionic moieties in the polymeric composition are provided by a blend of cationic polymer and anionic polymer, wherein each polymer is water soluble and film forming. Each polymer is generally

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formed by polymerisation of a monomer or monomer blend having the desired ionic character (anionic or cationic). If a blend is used, it may be a blend of ionic monomers of the chosen character or a blend with a non-ionic monomer.
5 Each monomer or monomer blend is usually water-soluble in order to ensure that the polymer is soluble in water.

The preferred cationic monomer, is diallyldimethyl ammonium chloride or other diallyl dialkyl ammonium salt. Other suitable cationic monomers include dialkylaminoalkyl-
10 (meth)acrylate or -(meth)acrylamide, either as acid salts or, preferably, quaternary ammonium salts, the alkyl groups may each contain 1 to 4 carbon atoms and the aminoalkyl group may contain 1 to 8 carbon atoms. Particularly preferred are dialkylaminoethyl (meth)acrylates,
15 dialkylaminomethyl (meth)acrylamides and dialkylamino-1,3-propyl (meth)acrylamides. These monomers may be present as acid salts or, preferably, quaternary ammonium salts. The dialkyl group is generally dimethyl, e.g., (meth)acrylamideopropyl trimethyl ammonium chloride.

20 To make cationic polymers for use in the invention the cationic monomers may be homopolymerised or copolymerised with each other or with a non-ionic monomer, preferably (meth)acrylamide to give a polymer formed of cationic moieties optionally with non-ionic moieties. Thus the
25 cationic polymers are usually formed from 30 to 100% by weight cationic monomer with the balance being non-ionic.

The amount of cationic monomer is usually at least 50% and often at least 80% by weight and most preferably 100%. Other suitable cationic polymers are polyethylene imines
30 and polyamines, e.g., polyamine epichlorhydrin polymers.

The molecular weight of the cationic polymer should preferably be such that the polymer has intrinsic viscosity of at least 0.1, and preferably at least 0.3 dl/g (measured by suspended level viscometer at 25°C in molar aqueous
35 sodium chloride). IV can be up to, for instance, 10dl/g or higher but is generally below 7, most usually below 4 and preferably below 2.5dl/g. Preferred values, especially for

polymers of diallyl dimethyl ammonium chloride, are in the range 0.8 to 2.4 dl/g. Such values indicate that preferred molecular weights in the region of around 50,000 to 1,000,000 or 2,000,000 and are preferably above 200,000.

5 If the molecular weight is too high, the forming composition may have an unacceptably high viscosity at reasonable concentrations of polymer.

The anionic monomers that can be used in the invention include acrylic acid, methacrylic acid, maleic acid, 10 2-acrylamido-2-methyl propane sulphonic acid (AMPS), sodium vinyl sulphonate, or other ethylenically unsaturated carboxylic or sulphonic acids, often as alkali or other water soluble salts.

To make anionic polymers for use in the invention, 15 anionic monomers may be copolymerised with each other or with non-ionic monomer such as (meth) acrylamide.

The proportions of anionic and nonionic monomer and molecular weights for the anionic polymers may all be similar to those discussed above for the cationic monomers, 20 although it can be desirable for the molecular weight to be a little higher, for instance 500,000 to 10 million, preferably 1 million to 5 million.

When the composition is based on an amphoteric polymer, the molecular weight may be as discussed above for 25 cationic polymers. The amount of non-ionic monomer in the blend is generally 0 to 50%, preferably 0 to 20%, by weight of the total monomer blend.

The polymers must be totally soluble in water. Accordingly, the polymers are preferably linear 30 homopolymers or copolymers of monoethylenically unsaturated water soluble monomers. However, cationic polymers which have a small degree of cross-linking or branching, or that are formed from monomers that include a small proportion of insoluble monomer, can also be suitable provided they do 35 not significantly affect solubility.

We have found that compositions formed from either ionic type alone do not have satisfactory properties for

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water soluble capsules but the use of a blend of ionic types, and preferably the use of a blend of counterionic polymers, gives the opportunity of obtaining a variety of properties that can be selected according to selection of the ratio of cationic and anionic moieties.

For instance, polymers of polyvinyl pyrrolidone or polyacrylamide or homopolymers of the anionic moieties described above tend to be too brittle when exposed to ambient humid atmospheres. Polymers of polyvinyl alcohol or homopolymers of the cationic moieties described above tend to have adequate flexibility but to be too sticky when exposed to ambient humid atmospheres. This problem of deliquescence applies also to some of the anionic polymers, and is particularly serious in respect of capsules of, for instance, cosmetic preparations such as bath oils.

By selecting appropriate proportions of the anionic and cationic monomers (i.e., moieties) it is possible, for the first time, to provide a synthetic shell that has the optimum combination of flexibility and other properties when exposed to an ambient atmosphere and yet which has a desired, usually very fast, rate of dissolution in water. If the amount of cationic monomer is too low then the shell will tend to be too brittle. If it is too high then the shell will tend to be too sticky in the presence of atmospheric humidity.

The molar ratio cationic:anionic moieties is preferably about 5:1 to 1:1, most preferably about 4:1 to 2:1.

The weight ratio cationic:anionic monomers is generally at least 1:1 and can be up to 8:1, but 4:1 is often sufficient. A preferred range is 1.5:1 to 3:1, with best results generally at around 2:1. These values are particularly preferred when the cationic polymer is based on diallyl dimethyl ammonium chloride (preferably as a homopolymer) typically having molecular weight in the range 300,000 to 1.5 million, generally 500,000 to 1,000,000 and when the anionic polymer is based on sodium acrylate or

methacrylate (generally as homopolymer) typically having molecular weight in the range 10,000 to 5,000,000, preferably around 50,000 to 2,000,000.

5 The shell-forming composition of the invention generally contains the defined synthetic polymer or polymer blend as the only film forming components. The composition is normally free of natural polymers such as gelatin.

10 An advantage of the invention is that the film can be formulated wholly from reproducible synthetic polymers. Another advantage is that it can be formulated to dissolve rapidly and completely, without leaving a scum in the water in which it is dissolved.

15 Another advantage of the invention is that the films can be heated, for instance during drying or formation of the capsules, without adversely affecting the properties of the film. This is in contrast to gelatin since that is liable to become brittle if it is heated significantly.

20 The capsule shell can be used to contain a variety of substantially anhydrous fillings. The filling must be substantially anhydrous otherwise the water in it may weaken the shell and may even result in rupture of the shell. Generally the water content of the filling is, initially, below 10% and preferably below 5% and, in particular, the total amount of water that is introduced by
25 the filling must be such that the resultant moisture of the film is sufficiently low, generally below 30%, preferably below 20% and most preferably below 10% even when exposed to an ambient atmosphere.

30 An advantage of the invention is that the film can easily be formulated to maintain structural integrity even when it does contain these small amounts of moisture.

The filling for the capsule can be liquid or powder and can be for instance selected from for instance
35 detergents, fabric softeners, fragrances, flavour oils, hazardous chemicals such as pesticides, herbicides or other agrochemicals, pharmaceuticals or cosmetics. It is

particularly preferred that the filling is a cosmetic, in particular bath oil or bath grains. If the filling is a solid, it will be in particulate form which can, typically, be such small particle size that, if it was not
5 encapsulated, might create a dusting hazard.

In a particularly preferred aspect of the invention a shell is prepared that is suitable for forming a capsule for containing a non-aqueous bath additive such as bath oil or bath grains characterised in that the capsule comprises
10 a water-soluble synthetic film-forming polymeric composition of cationic and anionic moieties in which the cationic moieties are provided by a cationic polymer and the anionic moieties are provided by an anionic polymer and the weight ratio of cationic to anionic polymer is from 1:1
15 to 8:1, preferably from 1.5:1 to 3:1, and most preferably around 2:1. The preferred polymers are polydiallyl dimethyl ammonium chloride and sodium polyacrylate or methacrylate, having the preferred molecular weights indicated above.

20 The shell capsules may be of any size depending on the requirement of the particular use. For example, for pharmaceutical preparations the capsules must be small enough for swallowing and may therefore be below 1cm diameter, even below 0.5 cm diameter. Preferably the
25 capsules have a diameter above 0.2cm, less preferably they could be microcapsules. For use as containers for bath oil, slightly larger capsules, such as those having diameter of from 1cm to 5cm may be preferred.

The compositions can contain, for instance, dyes
30 and/or pearling agents.

Examples

A variety of polymeric films suitable for making capsules were formed from various polymer solutions by drying, and the resulting films were evaluated for
35 characteristics of appearance and stability of films, water-solubilities at temperatures of from 35-45°C, mechanical strength, and resistance to non-aqueous oils.

The results are given in Tables 1 and 2.

TABLE 1

5	POLYMER	POLYMER FILM TYPE	WEIGHT RATIO	RATE OF DISSOLUTION AT 43-45°C (g/sec)
10	A	Poly DADMAC	-	0.005
	B	Poly DADMAC/ Polysodium acrylate + Pearling agent	3:1 3:1	0.009 0.003
15	C	Poly DADMAC/ Polysodium acrylate + Pearling agent	2:1 2:1	incomplete solubility -
	D	PolyDADMAC/ Poly Sodium Methacrylate	3:1	0.003
20	E	PolyDADMAC/ Poly Sodium Methacrylate	2:1	0.004
	F	PolyDADMAC/ Poly acrylic acid	2:1	0.007

TABLE 2

	POLYMER	MECHANICAL STRENGTH			
		60% REL.	HUMIDITY	30% REL.	HUMIDITY
5		Elongation At Break Point	Max.Load At Break Point	Elongation At Break Point	Max Load Load at Break Point
10	A	X 2	12N	X 1/2	150N
	B	X 2	20N	X 1/3	84N
	B + Pearling Agent	1/7	4N	X 1/10	69N
15	C	X 1/7	6N	X 1/3	107N
	C + Pearling Agent	1/7	3N	X 1/10	60N
20	D	X 2	16N	X 1/3	150N
	E	X 2	12N	X 1/3	110N
25	F	X 3	3N	X 1/10	98N

The mechanical strength tests were carried out at 22°C on polymeric film samples having thickness of from 0.4 to 0.5mm.

A subjective assessment was made of various films. PolyDADMAC having molecular weight around IV2 gives a tough opaque flexible film that is very sticky in humid atmospheres, whilst polyDADMAC having lower molecular weight forms a weaker film, that is again sticky in humid atmospheres. Polyacrylic acid (as the free acid) of molecular weight of around 2,000,000 gives a hard brittle film that is slightly sticky in a humid atmosphere whilst its sodium salt gives a brittle film that is slightly less sticky in humid atmospheres. Polyacrylamide gives an inflexible clear film.

Mixtures of two or three parts by weight polyDADMAC with one part by weight sodium acrylate or methacrylate homopolymer give flexible opaque films that are only slightly sticky when wet and which dissolve rapidly and fully in warm water. When the film is formed from two parts polyDADMAC and one part polyacrylic acid homopolymer (as free acid) the film is rather brittle.

Capsules may be formed for instance from the exemplified polymer compositions or other compositions of the invention by conventional capsule forming methods, including sachet folding or casting. For example using casting, the polymer solution may be cast into semi-spherical shapes and this is followed by sealing two semi-spheres together, for example using aqueous based adhesive or by moistening the edges of the semi-spherical shells and sealing them together by drying.

Capsules can also be prepared by having for example a semi-sphere of water-soluble shell adhered to a non water-insoluble hemisphere. Thus, the capsules do not have to be entirely water-soluble, but may comprise a wall which is partly formed of the water-soluble synthetic film-forming polymeric composition according to the invention.

The non-aqueous filling can be filled inside the shell capsules by known techniques, for instance injection into a fully formed, sealed shell capsule or by incorporation into the capsule during casting or other manufacturing step.

CLAIMS

1. A polymeric composition suitable for forming capsule shell for containing a substantially non-aqueous substance characterised in that the composition comprises a water soluble synthetic film-forming polymeric composition of cationic and anionic moieties.
2. A composition according to claim 1 in which the cationic moieties are selected from diallyl dialkyl ammonium salts, dialkylaminoalkyl (meth) acrylates and dialkylaminoalkyl (meth) acrylamides and the anionic moieties are selected from ethylenically unsaturated carboxylic or sulphonic acids.
3. A composition according to claim 1 in which the cationic moieties are selected from diallyl dimethyl ammonium chloride and dialkylaminopropyl (meth) acrylamides as acid addition or quaternary ammonium salts, and the anionic moieties are selected from acrylic acid, methacrylic acid and water soluble salts thereof.
4. A composition according to any preceding claim in which the weight ratio of cationic moieties to anionic moieties is from 1:1 to 8:1, preferably 1.5:1 to 4:1.
5. A composition according to any preceding claim which is a blend of a water soluble film-forming cationic polymer with a water soluble film-forming anionic polymer.
6. A composition according to claim 5 in which the cationic polymer is formed from 50 to 100% by weight cationic monomer and 0 to 50% by weight non-ionic monomer, and the anionic polymer is formed from 50 to 100% by weight anionic monomer and 0 to 50% by weight non-ionic monomer.
7. A composition according to claim 5 or claim 6 in which the cationic polymer has molecular weight 200,000 to 2 million and the anionic polymer has molecular weight 500,000 to 10 million.
8. Use of a composition according to any preceding claim for forming all or part of a capsule shell.
9. A capsule shell part or all of which is formed of a composition according to any of claims 1 to 7.

10. A filled capsule having a shell part of all of which is formed of a composition according to any of claims 1 to 7, and wherein the filling in the capsule is substantially non-aqueous.
- 5 11. A filled capsule having a shell formed of a composition according to any of claims 5 to 7 and containing as filling a non-aqueous bath additive selected from bath oil and bath grains.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 91/00930

I. CLASSIFICATION F SUBJECT MATTER (If several classification symbols apply, indicate all) ⁶ According to International Patent Classification (IPC) or to both National Classification and IPC Int.C1.5 A 61 K 9/48 A 61 K 9/50																														
II. FIELDS SEARCHED <div style="text-align: right; margin-right: 50px;">Minimum Documentation Searched⁷</div> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 30%; padding: 5px;">Classification System</td> <td style="padding: 5px;">Classification Symbols</td> </tr> <tr> <td style="padding: 5px;">Int.C1.5</td> <td style="padding: 5px;">A 61 K</td> </tr> </table> <div style="text-align: center; margin-top: 10px;">Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched⁸</div>			Classification System	Classification Symbols	Int.C1.5	A 61 K																								
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Y	---	8,9																												
<div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>¹⁰ Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&" document member of the same patent family</p> </div> </div>																														
IV. CERTIFICATION <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; padding: 5px;">Date of the Actual Completion of the International Search <div style="text-align: center;">02-09-1991</div></td> <td style="width: 50%; padding: 5px;">Date of Mailing of this International Search Report <div style="text-align: center;">0 3. 10. 91</div></td> </tr> <tr> <td style="padding: 5px;">International Searching Authority <div style="text-align: center;">EUROPEAN PATENT OFFICE</div></td> <td style="padding: 5px;">Signature of Authorized Officer <div style="text-align: center;">Falk Heck </div></td> </tr> </table>			Date of the Actual Completion of the International Search <div style="text-align: center;">02-09-1991</div>	Date of Mailing of this International Search Report <div style="text-align: center;">0 3. 10. 91</div>	International Searching Authority <div style="text-align: center;">EUROPEAN PATENT OFFICE</div>	Signature of Authorized Officer <div style="text-align: center;">Falk Heck </div>																								
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III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		Relevant to Claim No.
Category *	Citation of Document, with indication, where appropriate, of the relevant passages	
X	FR,A,2436213 (L'OREAL) 11 April 1980, see page 1, lines 1-5; page 3, lines 11-18; page 4, line 18 - page 5, line 8; page 25, example 72 ----	2-7
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